N64-17582

Code-1

FINAL REPORT

UNPUBLISHED PRELIMINARY DATA

NATIONAL AERONAUTICS AND SPACE ADMINISTRATION

NASA

~~RESEARCH~~ GRANT NsG 376

(NASA CR-53198) OTS: ---

Theoretical and Experimentsl Studies of the Underlying

Processes and Techniques of Low Pressure Measurement Final Report

under the direction of

D. Alpert 31 Jan. 1964 21 p refs

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1186022

Urbana, Illinois

January 31, 1964

OTS PRICE

XEROX $ 2.60 ph

MICROFILM $ 0.83 mf

I. Summary

During the past year, the program in low pressure measurement centered about two major activities: first a program of laboratory research in the general area of problems and techniques of low pressure measurement; second a survey of current activities in aeronomy, particularly those involved with experimental studies of the neutral density and composition of the upper atmosphere. The two phases of the overall effort were clearly related, since the scientific objectives in the experimental program were established with the field of aeronomy in mind; in fact, it was planned with a serious attempt to view the problems of low pressure measurement from a perspective which took into account NASA space activities as a whole.

The experimental program, which included a careful investigation of a number of methods for the measurement of ultrahigh vacuum, selected for special study certain anomalous effects which take place in ionization gauges in the presence of some molecular gases, notably oxygen. In a paper entitled "Effects of Electron-Surface Interactions in Ionization Gauges", the results of this study were presented at the recent Symposium of the American Vacuum Society. It was demonstrated that anomalous effects previously reported by Redhead (Vacuum 12, 267, 1962) were due to ionization by electron impact of gases adsorbed at the surface of the electrodes, in this case on the surface of the molybdenum grid of a Bayard-Alpert ionization gauge. Similar effects have also been observed with certain mass spectrometers.

The significant features and results of this study are described in the above paper, a preprint of which is attached. The results and

their interpretation are of particular significance in the measurement of neutral densities in the upper atmosphere, because very substantial anomalous effects are found in the presence of oxygen, at pressures precisely corresponding to those at which important satellite measurements have been made. As a result, very considerable interest in these results has been shown by workers in virtually every laboratory engaged in direct aeronomic measurement.

A detailed understanding of this phenomenon is of importance because it may explain certain anomalous results which have been observed in some satellite measurements, either of total or partial pressures. An interesting side light is that there may be significant implications of this work in a number of other fields unrelated to the measurement of pressure. These include certain high energy experiments, thermonuclear experiments and experiments in many fields of surface physics and chemistry. With respect to measurements in the upper atmosphere, implications of the effect and its understanding can be seen not only in satellite measurements at high altitudes but also in rocket measurements in the ionosphere. In addition to the special study described above, a continuing study of new and improved methods of measuring gas density was carried out. Included in this study were a comprehensive evaluation of the suppressor ionization gauge developed at the Coordinated Science Laboratory and a comparison of such gauges with other pressure measuring devices. A similar comparison and evaluation of various types of mass analyzers is also part of a continuing program. A general review of the state of the art in the field of ultrahigh vacuum constitutes a somewhat informally stated objective of the

Laboratory, an effort supported not only by this but also by other sponsors. As part of this objective, a study was also made of the present status of low temperature electron sources; clearly any major advance in this area would have profound implication in the instrumental techniques for the measurement of pressure. An invited paper entitled "Ultrahigh Vacuum" A Survey" was presented at the annual meeting of the American Physical Society and published in Physics Today 16, 22 (1963).

The survey of current activities in aeronomic experiments must be considered to be, at least in large measure, an effort on our part to become directly acquainted with the many efforts in this field. The portion of the report devoted to this topic should be viewed not as a final summary but as a progress report of a continuing effort. It soon became evident in the course of this study that relatively little of the most current and pertinent information was available in the published literature. In some cases, efforts to investigate recent flight experience could not be carried through because the experimenters themselves were still processing data. In other cases, where unsuccessful or questionable flights had been reported, it was virtually impossible to obtain clearcut explanations of the difficulties. This meant that an appraisal of the status of the field could only be approached via personal visits and discussions with experimenters in the field. Of great help to an over-all perspective was the Conference on Direct Aeronomic Measurements in the Lower Ionosphere held at the University of Illinois in October, 1963. The significant contribution of this get-together suggests that other conferences in this particular area would be of great value. It is probably most pertinent to observe that the state of the field as a whole is not at a point where a final appraisal of the various techniques is yet possible; it may be that this observation in itself is the most significant editorial comment which can be made at the present time.

II. Experimental Program

A. Studies of Anomalous Effects in Ionization Gauges

The attached paper by Schuemann, Segovia and Alpert describes the results of a major study which demonstrated and explained certain significant anomalous readings of ionization gauges when in the presence of some molecular gases, particularly oxygen and carbon monoxide. These results have been presented at the Symposium of the American Vacuum Society.

The important conclusions of this work are:

1. It has been shown that large anomalous ion currents may flow to the collector of an ionization gauge and give an indication which is unrelated to the actual gas density in the volume of the gauge enclosure. Such currents can be very sizeable, particularly when the gauges are operated in an oxygen atmosphere — at densities corresponding to those found at typical satellite and sounding rocket altitudes;
2. The anomalous currents give a false pressure indication which decays with time when the gauge is removed from the oxygen or vice versa. Such readings could easily be misinterpreted as an outgassing of the gauge;

3. Evidence has been provided for the interpretation of the effect as due to dissociative ionization by electron impact at the grid surface. At low electron currents there is an enhanced rate of production of such ions accompanied by a reduced rate of collection in a Bayard-Alpert gauge;
4. After exposure to oxygen at 10^{-7} Torr, anomalous currents corresponding to pressures of approximately 1.0×10^{-7} Torr have been observed with an emission current of a few milliamperes. Following the removal of oxygen, the effect decays with a time constant which varies inversely with the electron emission current. (At 1 ma emission current, the observed decay rate was approximately 10 minutes; at 10 ma, it was of the order of one minute.) In some of the Explorer XVII results, readings were taken less than 1 sec after increased exposure to the atmosphere. Under such circumstances a possible anomalous effect would not have time to decay to a small value and this might explain certain heretofore unexpected effects in the Explorer results;
5. In the presence of oxygen a number of other effects may also be present to give erroneous readings either in ionization gauges or in mass analyzers; in particular, the composition of the gas may be altered, both by chemical reactions at the hot filament and by electron bombardment of adsorbed gases.

This research program has greatly clarified some important effects which have significant bearing on pressure measurement in general. Among the topics which remain for further investigation are:

1. Investigation of the specific physical mechanisms by which ionization at the surface takes place for various gases and metals;
2. Further study of the desorption of gases by electron bombardment;
3. What are the mechanisms by which oxygen is converted into CO in an ionization gauge?
4. With a knowledge of the nature of the effect, is it possible to design an ionization gauge which does not exhibit the effect?
5. What is the best type of pressure measuring device for upper atmosphere research? (Naturally there may not be a unique answer to this question since the boundary conditions may vary greatly from experiment to experiment.)

A continuing program of research in these areas is of very considerable interest in many areas of research in the space science field.

B. Studies in New Methods of Pressure Measurement

During the past year, a number of studies have been carried out to evaluate and compare the reliability and limitations of a number of total and partial pressure manometers. One study involved a comparison between a magnetic deflection instrument (Davis and Vanderslice, 1960 Vacuum Symposium Transactions, Pergamon Press, 1961, p. 417) and a Philips omegatron (A. Klopfer and W. Schmidt, Vacuum 10, 363, 1960). Both were calibrated and compared with a Bayard-Alpert gauge in the region 10^{-5} to 10^{-10} Torr. In general, reasonable agreement was obtained; however, under certain conditions which will be under continuing investigation, the cracking patterns for molecular gases (O_2, CO) were found to vary with pressure in the magnetic deflection instrument, an effect which may be caused by surface ionization by electron impact. Hence further studies are called for. Modifications now being carried out on the Davis and Vanderslice instruments are expected to reduce greatly the rather large outgassing rates observed, especially upon first starting a given experimental run.

In general, both the Davis and Vanderslice instrument and the omegatron require an experienced operator, with a number of ideosyncracies in each instrument which can only be learned after considerable experience. Of all of the pressure measuring instruments thus far used, the omegatron interacts the least with the gases in the system and seems to give the most reliable results. However, this series of comparisons did not include a Paul quadrupole mass filter, a type of instrument with which we expect to experiment in the continuing program.

During the year, considerable experience has built up in the use of the photocurrent suppressor gauge, invented at the Coordinated Science Laboratory during 1962 (W. C. Schuemann, Rev. Sci. Instr. 34, 700, 1963). This gauge greatly reduces the x-ray photocurrent limitations of the Bayard-Alpert gauge, while retaining many of its advantages. Linearity and reliability were demonstrated in the region of pressures from 10^{-5} to 5×10^{-12} Torr.

Considerable experience was also built up in the properties of the modulator gauge (Redhead, Rev. Sci. Instr. 31, 343, 1960) and its characteristics and limitations analyzed. References to some of these are described in the attached reports.

C. Thin Film Electron Sources

The presence of a thermionic cathode in mass spectrometers and ion gauges frequently alters the identity of the gases measured by these instruments. A simple and robust cold electron source inert with respect to its surroundings would be very desirable. When Mead (J. Appl. Phys. 32, 646, 1961) suggested the use of thin film multi-layer devices as electron sources, it looked as though a solution of this problem might be at hand. A suitable source for use in modern vacuum techniques should not be damaged by bakeout at temperatures up to at least 400°C, nor should it contribute "dirt" to the system at these temperatures. For many applications a long, stable life would be a necessity.

Mead sources are three-layer devices consisting of an evaporated metal base, a thin insulating film a few hundred angstroms thick, and a thin covering metal film operated a few volts positive with respect to the base. Initial attempts to fabricate such sources in the Coordinated Science Laboratory were successful to the extent that electron emission was observed. The usual materials of the sandwich were aluminum-aluminum oxide-gold, although tin oxide was also used with several metals. It should be noted that at the insulator thicknesses used in this Laboratory and also for those used by Hickmott (J. Appl.

Phys. 33, 2669, 1962) and others, the spacing between the metal layers is so great that the "diode" current in the insulator is not the result of quantum mechanical tunneling. Therefore, the frequently used name, "tunnel cathode", for a source of the present type is a misnomer unless the insulator is less than about 50 Å thick. The conduction process in the thicker films is complicated and not well understood.

Electron emission currents up to several microamperes were observed in samples prepared under the present contract. The operation of a source at such currents gave at best fractional-hour life. In fact, merely bringing the device to operating voltage (about 10-12 volts) caused irreversible changes in the diode conductivity of the sample. The emission current for all samples, of any materials tried, was "noisy". A source of a few mm^2 area was placed several centimeters from a phosphor screen to permit examining a magnified image of the emission pattern. Most of the electrons were found to originate from a number of spots that flickered on and off, the number of spots remaining approximately constant. A small steady current component was seen.

Much better cold cathodes have been made by Cohen (J. Appl. Phys. 33, 199, 1962 and Appl. Phys. Ltrs. 1, 61, 1963). Cohen used cesium to lower the work function of the outer metal film and thus increased the emission of his sources by 10^5. This technique could be very useful for a permanently evacuated tube, but it would not be applicable to one baked-out or opened to air after sensitization, or to one operated under even moderate pressures of active gases.

Perhaps the most useful electron source could be made by combining a Mead-type cathode with a resistance strip electron multiplier. By operating the film device at very low currents it would be more reliable, and a multiplier could increase the current to a value of at least several microamperes.

III. Survey of Measurement of Neutral and Ion Composition of the Upper Atmosphere

One of the most important and challenging research efforts in the field of aeronomy at the present time is the measurement of the neutral and ion composition of the upper atmosphere. In the following section we shall restrict ourselves to the discussion of neutral composition measurements. The problem of the measurement of ion composition will be treated in a subsequent report.

The first attempt to measure the atmospheric composition with a mass spectrometer was made by Townsend and co-workers (Rocket Exploration of the Upper Atmosphere, edited by Boyd, Seaton and Massey, Inter-Science Publishers, 1954) in 1953. A Bennett mass spectrometer (Townsend, Rev. Sci. Instr. 23, 538, 1952) was flown in an Aerobee rocket over White Sands with the objective to measure diffusive separation of atmospheric constituents by monitoring the N_2/A ratio as a function of height. No diffusive separation was detected, but a number of error sources made the interpretation of the data rather unreliable. Despite this shortcoming, it was the first demonstration of the use of a mass spectrometer in a rocket experiment. An improved Bennett spectrometer was flown by the same group in 1956 and 1958 over Fort Churchill

(Townsend, et al., A. of the IGY 12, 431, 1960). In these flights evidence for diffusive separation at altitudes of 120 km was obtained, as well as qualitative data on the upper atmospheric composition.

Around the same time (1959), similar experiments were conducted in the Soviet Union by Pokhunkov (Planet. Space Sci. 9, 269, 1962; Planet. Space Sci. 11, 441, 1963), who also used a Bennett spectrometer. These flights revealed a number of typical difficulties that are encountered in the analysis of the neutral atmosphere. The following error sources can be identified:

1. No precautions were taken to discriminate against backstreaming gas entering the ionization region and becoming ionized. Consequently, it is very difficult to determine the true ambient pressure from the ion currents;
2. As a direct consequence of (1), sizeable contributions from outgassing were recorded. Some recorded mass numbers like H_2O, CO_2 and NO were attributed entirely to outgassing and secondary reactions taking place in the spectrometer;
3. No precautions were taken to prevent atomic oxygen from hitting surfaces prior to entering the ionization region. Hence, a substantial recombination of atomic oxygen might have occurred;
4. The use of low energy ionizing electrons (36 eV) introduces a considerable uncertainty in the absolute values of the neutral concentrations, because the ionization cross sections are very sensitive to energy in this range.

The use of low energy ionization energies is, of course, motivated by the desire to prevent multiple ionizations. It may be questionable, however, if the uncertainty in the ionization probability is not larger than the contribution of doubly ionized ions, a contribution which, moreover, can be taken into account.

The remaining part of this section will be devoted to a discussion of current efforts on the mass spectrometric measurements of neutral composition. Since most of these efforts are not yet available in the open literature, visits were arranged to the main laboratories that are actively engaged in this field or, in some cases, private discussions were conducted with scientists working in this area.

A. High Altitude Engineering Laboratory, Department of Aeronautical and Astronautical Engineering, University of Michigan. E. J. Schaefer

This group started to develop a mass spectrometer for use in sounding rockets in 1958. The instrument is a quadrupole mass spectrometer (Paul, et al., Z. fur Naturf. 8a, 448, 1953; Paul and Raether, Z. fur Physik 140, 262, 1955; and Paul, et al., Z. fur Physik 152, 143, 1958) designed to measure the neutral composition at altitudes above 100 km where no differential pumping is required. The quadrupole mass spectrometer was given preference over a magnetic one because it is lighter, simpler in construction, easier to incorporate into a payload and within generous limits insensitive to pressure and initial energy of the ions.

The main parameters of the instrument are:

Length: 12.75 cm and 17.8 cm

Field radius: 0.52 cm

Aperture: 0.081 cm diameter with an acceptance angle of 5.25^{o}

Frequency: 2.39 mc

rf voltage: 500 V at mass 40

Resolution $M/\Delta M = 40$

rf power: 3 watts

The ion source operates with an electron emission current of 0.4 and 4 ma at 45 V. 45 V is also the acceleration potential for the ions. The filaments are rhenium; the electrical structure of the ion source is gold-plated to reduce as much as possible the recombination of atomic oxygen.

The separated ion current is detected with an electrometer of 10^{-12} amp sensitivity at a 5 ms time constant. In conjunction with the ion source this corresponds to a partial pressure of 10^{-7} Torr. The scan rate is 1/sec and 2/sec. Two modes of scanning are employed. In the first mode, the rf voltage is varied, keeping the ratio of dc-to-ac voltage, and thus the width of the stable region constant. This results in a line-type mass spectrum. In the second mode, the dc voltage is switched off. This results in an integrated mass spectrum in the sense that for a given rf voltage all masses greater than M_o reach the collector.

In order to keep contamination from the rocket to a minimum, the mass spectrometer package is ejected from the rocket at an appropriate altitude. Most of the previous failures during firings were due to a malfunctioning of this ejection mechanism rather than to a failure of the mass spectrometer itself.

To date, two successful flights have been performed in 1962 and 1963 (Schaefer and Nichols, COSPAR IV. International Space Science Symposium, Warsaw, Poland, June, 1963; Schaefer, J. Geophys. Res. 68, 1175, 1963). In the first flight data were collected on the neutral composition for 100 - 130 km; in the second flight, the range was increased to 190 km. For the evaluation of the data on the atomic oxygen concentration it was assumed that the cross section for ionization of atomic oxygen is the same as that for molecular oxygen (Fite and Brackmann, Phys. Rev. 113, 815, 1959; Rothe, *et al.*, Phys. Rev. 125, 582, 1962). Some uncertainty existed as to the exact energy of the ionizing electrons. There was reason to suspect that the actual electron energy was somewhat lower on 45 V due to space charge effects. The contribution of dissociative ionization of O_2 to the O^+ ion current was determined experimentally in the Laboratory, and a value of four percent O^+ from O_2 for 45 V was obtained. In view of the low electron energy of 45 V and the uncertainty in this value, the comments made earlier on the use of low electron energies for the ion source apply.

B. Space Physics Laboratory, Department of Electrical Engineering, University of Michigan. G. R. Carignan, Nagy, Niemann, Taeusch

Among various other projects that lie outside the scope of this survey, a very interesting method has been designed to measure the temperature of the neutral gas in the upper atmosphere.

This method uses an omegatron tuned to N_2. From the shape of the mass-line, the temperature of N_2 can be deduced. The same information could, of course, be obtained from the line shapes in magnetic deflection instruments and actually reaction energies between molecular species have been measured in this way, but this would be much more difficult in a rocket experiment than by using an omegatron. Since only one mass number is monitored, the usual difficulties associated with the use of omegatrons as mass spectrometers or partial pressure analyzers are to a great extent eliminated.

It is planned to use the omegatron, however, for the measurement of neutral gas composition in the lower atmosphere, but no flights have been performed so far.

C. Bell & Howell Research Center, Pasadena, California. W. M. Brubaker

Two quadrupole mass spectrometers have been developed which will be referred to as BH I and BH II. BH I is a small spectrometer designed for A. F. C. R. L. in cooperation with R. Narcisi for the measurement of neutral and ion composition in the region from 70 km up. The high ambient pressure in the D region necessitates the use of differential pumping techniques to maintain a low enough pressure in the mass spectrometer ($\sim 10^{-4}$ Torr). This is achieved by the use of a liquid nitrogen-cooled zeolite trap that acts as a cryogenic pump with an equivalent pumping speed of the order of 100 liters/sec at 10^{-4} Torr and 50 liters/sec at 10^{-3} Torr.

The instrument itself has the following characteristics:

Length: 7.6 cm

Field radius: 0.38 cm

Aperture: 0.038 cm diameter

rf voltage: 310 V at mass 44

Frequency: 6 mc

rf power: 4.5 watts

Acceleration voltage: 125 V

Resolution: 10% between peaks at mass 16

Ion source: Current 0.5 ma; Voltage 235 V

The choice of a higher acceleration voltage for the ionized electrons makes the cross section for ionization much less sensitive to the electron energy than in Schaefer's design.

The electrometer has a logarithmic response from 10^{-12} to 10^{-7} amps with a time constant of 10 milliseconds. The ion source is designed to have a high sensitivity to incoming molecules, but a very low sensitivity to backstreaming gas. The instrument has had one successful flight so far.

BH II is a quadrupole mass spectrometer for use in satellites. It is 25.4 cm long and has a field radius of 1.52 cm. It is designed as an ion mass spectrometer; no ion source is provided in flight. No actual flights have been performed.

D. Consolidated Systems Corporation, Monrovia, California. L. Hall

Three different mass spectrometers for neutral composition measurement have been designed or are under design at CSC:

1. A double focusing magnetic spectrometer that is presently being flown on Explorer XVII;
2. A quadrupole spectrometer for use in sounding rockets;

3. A quadrupole spectrometer for the measurement of the martian atmospheric composition.

1. The double focusing instrument uses magnetic and electric deflection techniques in order to focus ions of different energy onto the collector. In this instrument transverse energies of 12 eV can be accepted. The ion source operates at a voltage of 45 V and 0.5 ma emission current. The minimum detectable partial pressure is of the order of 10^{-11} Torr. This high sensitivity is achieved through the use of long integration times in the electrometer. The scan rate is thus limited to 1/min which is short enough for satellites. Preliminary results for Explorer XVII indicated that excessive amounts of O_2 were detected, an effect which was tentatively attributed to an outgassing of the titanium electrodes used in the ion source.

2. The quadrupole spectrometer is still in the developmental stage. The main characteristics are:

Length: 20.2 cm

Field radius: 0.75 cm

Aperture: 0.05 cm diameter

rf voltage: 400 V

Frequency: 1.3 - 3.0 mc

Resolution: 1% between peaks at mass 20

Acceleration voltage: 20 V

The ion source has a sensitivity of 10^{-4} amp/Torr and operates with an emission current of 0.5 ma. Contrary to the previously described quadrupole spectrometers, this instrument utilizes a frequency scan rather than a voltage scan. The frequency is varied in steps, corresponding to discrete, preselected mass numbers. This requires that the rf voltage is kept constant during the scan and feedback regulation must be provided. Great care has been taken to assure that the mass peaks are flat-topped, so that abundance ratios can be measured reliably. This condition is realized for entrance angles of $\pm 3^{\circ}$.

3. The martian atmosphere instrument is similar to the previous one, except that it is six inches long and has a field radius of 0.5 cm. The discrete frequency scan has a great advantage for this model, because it results in a considerable saving in telemetry bandwidth, which is an important factor in the transmission of data over interplanetary distances.

E. Geophysics Corporation of America (GCA), Bedford, Massachusetts. R. F. K. Herzog, F. F. Marmo

A helium mass spectrometer is being developed which utilizes two 90° magnetic deflection sectors in order to reduce scattered ion currents and make the measurement of extremely low helium concentrations feasible. The objective of this instrument is to measure the helium-to-argon abundance ratio in order to determine accurately the diffusive separation. The instrument is to be flown in an Aerobee rocket, but no actual flight has been performed so far.

F. University of Minnesota, Minneapolis, Minnesota. A. O. C. Nier

A small double focusing magnetic mass spectrometer was developed at the University of Minnesota for possible use in rocket flights (Nier, *et al.*, Rev. Sci. Instr. 31, 1127, 1960). This instrument, together with a small 90° magnetic spectrometer, was flown successfully in June, 1963 (Nier, et al., Conference on Direct Aeronomic Measurements in the Ionosphere, October, 1963, Urbana, Illinois). A vac-ion pump is incorporated into the payload to provide differential pumping. Although the filament of the double focusing instrument failed in flight, the single focusing instrument gave a wealth of data. Although the results have not yet been published, the preliminary data presented at the Aeronomy Conference in Urbana indicated that these are probably the best data obtained so far in the range 95 - 200 km.

IV. Conclusions

As indicated in the Summary, this activity must be considered an attempt on the part of this Laboratory to become directly familiar with the many efforts in this field. Most of the activity in this direction consisted of reviewing the literature and making visitations to a number of the most active programs in this country's aeronomy effort. It was our experience that the visits and conversations had a beneficial effect well beyond the collection of information. Most of the scientists visited seemed to welcome the opportunity to exchange views as well as information and found it easy to do so with a group whose objectives were in no sense competitive or directly overlapping their own. Thus, in an important sense, an integration and exchange process seemed to be taking place in the course of this acquisition of information.

It is apparent from the previous discussion that a number of difficulties are encountered when mass spectrometers are used for the measurement of the neutral composition in rockets and satellites. Several of these difficulties can be avoided or eliminated by a careful design of the mass spectrometer and the payload. The measurements by Nier are probably a good example of the elimination of problems through careful engineering.

There are, however, more basic problems that require additional work in the Laboratory. Some of these are:

1. More data are needed on ionization cross sections, in particular for low energies and atomic species. Cross sections for double ionization are needed for almost all atmospheric constituents;
2. More information is needed on the interaction of atomic oxygen with surfaces;
3. Data on the interaction of high energy neutral molecules with surfaces should be obtained in order to get more reliable values for the accomodation coefficients;
4. Theoretical and experimental studies on the hypersonic flow around rockets and its influence on the sampling of neutral particles and ions are necessary for the proper interpretation of data at low altitudes.